

# Transport in multilevel quantum dots: from the Kondo effect to the Coulomb blockade regime

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A new theoretical method is introduced to study coherent electron transport in an interacting multilevel quantum dot. The method yields the correct behavior both in the limit of weak and strong coupling to the leads, giving a unified description of Coulomb blockade and the Kondo effect. Results for the density of states and the temperature dependent conductance for a two-level dot are presented. The relevance of these results in connection to recent experiments on the Kondo effect in semiconducting quantum dots<sup>1,2</sup> is discussed.

The proper description of coherent electron transport in the presence of strong electron-electron interactions has been one of the central issues in the field of mesoscopic systems<sup>3</sup>. Semiconducting quantum dots (QDs) provide an almost ideal system where the predictions of the theory can be tested and new effects could be searched for. For instance, recent experiments have demonstrated the possibility of exploring the Kondo effect, a prototypical correlation effect, using this technology<sup>1,2</sup>.

From the theoretical point of view, correlation effects have been mainly analyzed in this kind of systems by means of the simple Anderson model with a single spin-degenerate level<sup>4,5</sup>. Within this model the Kondo effect arises due to fluctuations in the spin of an unpaired electron<sup>6</sup>. So far, there has been few attempts to include a multilevel spectrum for describing either single QDs with quasi-degenerate levels or coupled QDs beyond a rate equation approach<sup>7</sup>. The actual situation in semiconducting QDs should require the inclusion of a multilevel spectrum whenever many single-particle states with a small level separation are involved in the transport process. This could be the case in experiments measuring the phase of the transmission amplitude through a QD in the Coulomb blockade (CB) regime<sup>8</sup> and also in recent experiments on the Kondo effect<sup>1,2</sup>.

The aim of this letter is to introduce a new theoretical approach for describing correlation effects in multilevel QDs. The approach is constructed to yield the correct behavior both in the limit of infinite and vanishing charging energy. This is achieved by introducing an interpolative self-energy for the one-electron Green functions, an approach which has been successfully applied to several interacting systems including the equilibrium and non-

equilibrium Anderson models<sup>5,9</sup>. This type of approach has been recently rediscovered and applied to analyze the Mott transition in the Hubbard model for arbitrary band filling<sup>10,11</sup>.

For describing the multilevel QD we consider a model Hamiltonian which is a generalization of the single-level Anderson model,  $H = H_{\text{dot}} + H_{\text{leads}} + H_T$ , where  $H_{\text{dot}} = \sum_m \epsilon_m \hat{d}_m^\dagger \hat{d}_m + U \sum_{l>m} \hat{n}_m \hat{n}_l$  corresponds to the uncoupled QD ( $\hat{n}_m = \hat{d}_m^\dagger \hat{d}_m$ );  $H_{\text{leads}} = \sum_{k \in L, R} \epsilon_k \hat{c}_k^\dagger \hat{c}_k$  to the uncoupled leads, and  $H_T = \sum_{m, k \in L, R} t_{m, k} \hat{d}_m^\dagger \hat{c}_k + h.c.$  describes the coupling between the dot and the leads. The labels  $m$  and  $l$  in  $H$  denote the different dot levels including spin quantum numbers.

Our main objective is the determination of the dot retarded Green functions from which the different level charges and the linear conductance can be obtained. In a frequency representation they can be written as  $G_m(\omega) = [\omega - \epsilon_m^{HF} - \Sigma_m(\omega) - i\Gamma_{m, L}(\omega) - i\Gamma_{m, R}(\omega)]^{-1}$ , where  $\epsilon_m^{HF} = \epsilon_m + U \sum_{l \neq m} n_l$  is the Hartree-Fock level (we adopt the notation  $n_l = \langle \hat{n}_l \rangle$ ),  $i\Gamma_{m, L(R)}(\omega) = \sum_{k \in L(R)} t_{m, k}^2 / (\omega - \epsilon_k + i0^+)$  are the hybridization terms describing the coupling to the leads and  $\Sigma_m(\omega)$  is a self-energy that takes into account electron-electron interactions beyond the Hartree-Fock approximation. We shall neglect the indirect coupling between dot states<sup>12</sup> and adopt the usual approximation of  $\Gamma_{m, L}$ ,  $\Gamma_{m, R}$  being independent of energy. In our approach<sup>5,9</sup>, we look for an interpolative self-energy yielding the correct  $U/\Gamma_m \rightarrow 0, \infty$  limits.

In the  $U/\Gamma_m \rightarrow \infty$  or “atomic” limit,  $G_m(\omega)$  can be obtained using the equation of motion method<sup>13</sup>, which yields

$$G_m^{(at)}(\omega) = \frac{\langle \prod_{l \neq m} (1 - \hat{n}_l) \rangle}{\omega - \epsilon_m + i0^+} + \sum_{l \neq m} \frac{\langle \hat{n}_l \prod_{(s \neq l) \neq m} (1 - \hat{n}_s) \rangle}{\omega - \epsilon_m - U + i0^+} + \dots + \frac{\langle \prod_{l \neq m} \hat{n}_l \rangle}{\omega - \epsilon_m - (M-1)U + i0^+}, \quad (1)$$

where  $M$  denotes the total number of one-electron levels in  $H_{\text{dot}}$ . In this expression all possible charge states of the dot give a contribution. Their evaluation requires

the knowledge of many particle correlations  $\langle \hat{n}_1 \hat{n}_2 \rangle$ ,  $\langle \hat{n}_1 \hat{n}_2 \hat{n}_3 \rangle$ , etc. However, for sufficiently large  $U$  fluctuations in the dot charge with respect to the mean

charge  $\mathcal{N}$  by more than one electron become negligible and  $G_m^{(at)}(\omega)$  is accurately given by the three poles expression

$$G_m^{(at)}(\omega) \simeq \frac{A_{N-1}^m}{\omega - \epsilon_m - U(N-1) + i0^+} + \frac{A_N^m}{\omega - \epsilon_m - UN + i0^+} + \frac{A_{N+1}^m}{\omega - \epsilon_m - U(N+1) + i0^+}, \quad (2)$$

where  $N = \text{Int}[\mathcal{N}]$ . In order to yield the exact first three momenta of Eq. (1) the weight factors  $A_N^m$  should satisfy the following sum rules

$$\begin{aligned} A_{N-1}^m + A_N^m + A_{N+1}^m &= 1 \\ (N-1)A_{N-1}^m + NA_N^m + (N+1)A_{N+1}^m &= \sum_{l \neq m} n_l \\ (N-1)^2 A_{N-1}^m + N^2 A_N^m + (N+1)^2 A_{N+1}^m &= \sum_{l \neq m} n_l + \langle \hat{n}\hat{n} \rangle_m, \end{aligned} \quad (3)$$

where  $\langle \hat{n}\hat{n} \rangle_m = \sum_{(l \neq k) \neq m} \langle \hat{n}_l \hat{n}_k \rangle$ . The special case  $N = 0$  ( $N = M - 1$ ) has to be treated as  $N = 1$  ( $N = M - 2$ ). Notice that this expression for  $G_m^{(at)}(\omega)$  is fully determined by the average charges  $n_l$  and the two-body correlations  $\langle \hat{n}_l \hat{n}_k \rangle$ . From  $G_m^{(at)}(\omega)$  one can define an “atomic” self-energy,  $\Sigma_m^{(at)} = \omega - \epsilon_m^{HF} - [G_m^{(at)}(\omega)]_m^{-1}$ . Using Eqs. (2) and (3) it can be shown that  $\Sigma_m^{(at)}$  can be written as the ratio of two polynomials in  $\omega$  of the form

$$\Sigma_m = \left( \frac{\Sigma_m^{(2)}}{\alpha_m} \right) \frac{a_m + (a_m \Delta \epsilon_m / U + b_m / \alpha_m) \left( \Sigma_m^{(2)} / U \right)}{1 + (2\Delta \epsilon_m / U + c_m) \left( \Sigma_m^{(2)} / \alpha_m U \right) + (c_m \Delta \epsilon_m / U + d_m) \left( \Sigma_m^{(2)} / \alpha_m U \right)^2}. \quad (6)$$

This expression provides the generalization to the multilevel case of the interpolative self-energy first introduced in<sup>9</sup> for the single level Anderson model. The single level case ( $M = 2$ ) is readily obtained from Eq. (6) when  $N = 1$  taking  $\langle \hat{n}\hat{n} \rangle_m = 0$ . It is evident by construction that  $\Sigma_m \rightarrow \Sigma_m^{(at)}$  when  $U/\Gamma_m \rightarrow \infty$  and  $\Sigma_m \rightarrow \Sigma_m^{(2)}$  in the small  $U$  limit.

The final step in this approach is to determine the level charges  $n_m$ , the correlation functions  $\langle \hat{n}_l \hat{n}_k \rangle$  and the effective levels  $\tilde{\epsilon}_m$  selfconsistently. The charges and the correlations functions are determined through the relations  $n_m = -\int_{-\infty}^{\infty} f(\omega) \text{Im} G_m(\omega) d\omega / \pi$  and

$$\sum_{l \neq m} \langle \hat{n}_l \hat{n}_m \rangle = \frac{-1}{\pi U} \int_{-\infty}^{\infty} f(\omega) \text{Im} [\Sigma_m(\omega) G_m(\omega)] d\omega. \quad (7)$$

Eq. (7) is an exact relation that follows from the equation of motion of the retarded Green functions. It is important to stress that the self-consistent determination of the two-body correlations is *essential* to get the correct values of the charge for large  $U$ . The effective level is determined by imposing the condition:

$$\Sigma_m^{(at)} = \frac{a_m U^2 (\omega - \epsilon_m + i0^+) + b_m U^3}{(\omega - \epsilon_m + i0^+)^2 + c_m U (\omega - \epsilon_m + i0^+) + d_m U^2}, \quad (4)$$

where  $a_m = (\mathcal{N} - n_m) [1 - (\mathcal{N} - n_m)] + \langle \hat{n}\hat{n} \rangle_m$ ;  $c_m = \mathcal{N} - n_m - 3N$ ;  $d_m = \langle \hat{n}\hat{n} \rangle_m + 3N^2 - 1 - (3N - 1)(\mathcal{N} - n_m)$  and  $b_m = N^2(1 - N) - (\mathcal{N} - n_m)d_m$ . On the other hand, in the  $U/\Gamma_m \rightarrow 0$  limit  $\Sigma_m$  is given by the second order expression<sup>9</sup>

$$\Sigma_m^{(2)}(\omega) = U^2 \sum_{l \neq m} \int_{-\infty}^{\infty} d\epsilon_1 d\epsilon_2 d\epsilon_3 \frac{\tilde{\rho}_m(\epsilon_1) \tilde{\rho}_l(\epsilon_2) \tilde{\rho}_l(\epsilon_3)}{\omega + \epsilon_2 - \epsilon_1 - \epsilon_3 + i0^+} [f_1 f_3 (1 - f_2) - (1 - f_1) (1 - f_3) f_2], \quad (5)$$

where  $f_i = f(\epsilon_i)$  denotes the Fermi distribution function at the leads, and  $\tilde{\rho}_m(\omega)$  are effective densities of states given by  $\pi \tilde{\rho}_m(\omega) = \Gamma_m / ((\omega - \tilde{\epsilon}_m)^2 + \Gamma_m^2)$ . The effective levels  $\tilde{\epsilon}_m$  are introduced in order to fulfill the Fermi-liquid relations associated with charge conservation (Friedel sum rule (FSR)<sup>14</sup>) as discussed below.

In order to determine an interpolative scheme between the two limits let us first notice that  $\Sigma_m^{(2)} \rightarrow U^2 \alpha_m / (\omega - \tilde{\epsilon}_m)$ , where  $\alpha_m = \sum_{l \neq m} \tilde{n}_l (1 - \tilde{n}_l)$ , when  $\Gamma_m / \omega \rightarrow 0$ . On the other hand,  $a_m \rightarrow \alpha_m$  in the small  $U$  limit and thus  $\Sigma_m^{(at)} \rightarrow U^2 \alpha_m / (\omega - \epsilon_m)$  in this case. These properties suggest to define the interpolative self-energy, replacing  $\omega - \epsilon_m$  by  $\alpha_m / \Sigma_m^{(2)} + \Delta \epsilon_m$ , where  $\Delta \epsilon_m = \tilde{\epsilon}_m - \epsilon_m$ , in Eq. (4) for  $\Sigma_m^{(at)}$ , which yields

$$\int_{-\infty}^{\infty} f(\omega) \text{Im} \left[ G_m(\omega) \frac{\partial \Sigma_m}{\partial \omega} \right] d\omega = 0, \quad (8)$$

which at zero temperature reduces to the Luttinger theorem<sup>15</sup> ensuring the fulfillment of the FSR<sup>14</sup>  $n_m = -\text{Im}[\ln G_m(E_F)] / \pi$ . In Ref.<sup>5</sup> we showed that the condition of consistency between the effective and the final charges is nearly equivalent to imposing the fulfillment of the FSR for the simple Anderson model. Similar self-consistency conditions have been proposed in Refs.<sup>10,11</sup>.

We have first applied this formalism to the case of a doubly degenerate level (four-fold degeneracy when including spin) which is a simple generalization of the single level Anderson model. The inset in Fig. 1 shows the charge per level  $n_m$  as a function of the leads Fermi energy  $E_F$ . For the case shown in Fig. 1, corresponding to  $\epsilon_m = 0$ ,  $\Gamma_m / U = 0.075$  (same for all levels) and zero temperature, one can observe a modulation in the charge resembling the typical Coulomb staircase of the  $\Gamma_m / U \rightarrow 0$  limit (shown as dotted line for comparison).

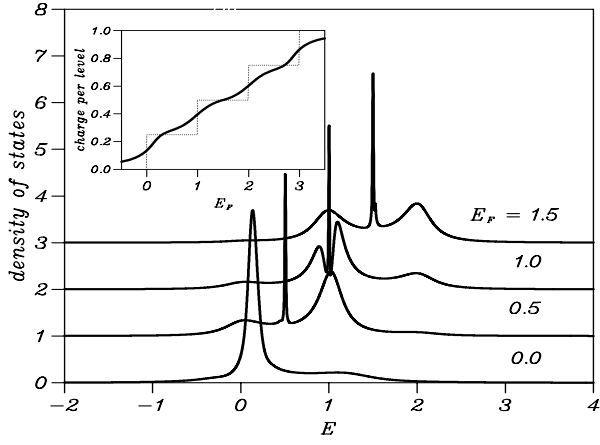


FIG. 1. Density of states for a fully degenerate two-level dot with  $\Gamma_m/U = 0.075$  and  $\epsilon_m = 0$  for different values of the leads Fermi energy  $E_F$ . The inset shows the charge per level and spin as a function of  $E_F$  (full line) as compared to the  $\Gamma_m/U \rightarrow 0$  case (dotted line). Energies are measured in units of  $U$ .

Fig. 1 also shows the density of states (DOS) associated with the dot levels for different values of  $E_F/U = 0, 0.5, 1.0, 1.5$  corresponding to the steps and the center of the plateaus in the charging curve. For  $E_F/U = 0$ ,  $n_m \simeq 0.125$  (the total charge on the dot being  $\sim 0.5e$ ) the system is in the so-called mixed-valence regime. The spectrum is in this case similar to the one found using large  $N$ -expansions in the  $U \rightarrow \infty$  limit<sup>16</sup> showing a resonance at a renormalized level just above  $E_F$ . For  $E_F/U = 0.5$  and  $1.5$  there are approximately one and two electrons respectively inside the dot. For these cases the system is in the Kondo regime, the DOS exhibiting a sharp peak around  $E_F$  (Kondo resonance) half way between two broader resonances (of width  $\sim \Gamma$ ) approximately separated by  $U$ , corresponding to the elementary charge excitation energy. It is worth noticing that the weight of these resonances is the same for the  $E_F/U = 1.5$  case due to the electron-hole symmetry in the half-filled dot, while there is a pronounced asymmetry for  $E_F/U = 0.5$ . For a half-integer occupation of the dot like in the  $E_F/U = 1.0$  case the DOS exhibits a more complex structure with three broad resonances around  $E = 0, U$  and  $2U$ , and a narrow Kondo peak still present at  $E_F$ . The overall shape is reminiscent of an average between the cases with one and two electrons in the dot.

The evolution of the Kondo peak as a function of  $E_F$  and temperature should be reflected in the dot linear conductance,  $G$ , which can be readily obtained from the Green functions (see Ref.<sup>17</sup>). The conductance as a function of  $E_F$  and different temperatures is shown in Fig. 2. When approaching zero temperature the conductance behaves like  $G = 4(e^2/h) \sin^2[\pi n_m]$  as expected from the FSR. The conductance decreases very rapidly with temperature in the region  $0.5 < E_F < 2.5$  where the DOS at  $E_F$  is controlled by the Kondo peak. Outside this region one can notice a slight increase of conduc-

tance with temperature. At temperatures large enough to be above the Kondo temperature,  $T_K$ , which can be estimated by the condition  $G(T_K) \simeq G(0)/2$ <sup>1</sup>, the conductance tends to exhibit the usual CB peaks at the charge degeneracy points. We should remark that our approach does not provide an accurate estimate of  $T_K$  as a function of the model parameters as the exponential decrease of the Kondo peak weight for very large  $U$ <sup>18</sup> is not strictly recovered. This limitation does not affect, however, the qualitative behavior of the conductance except for a rescaling of the temperature values.

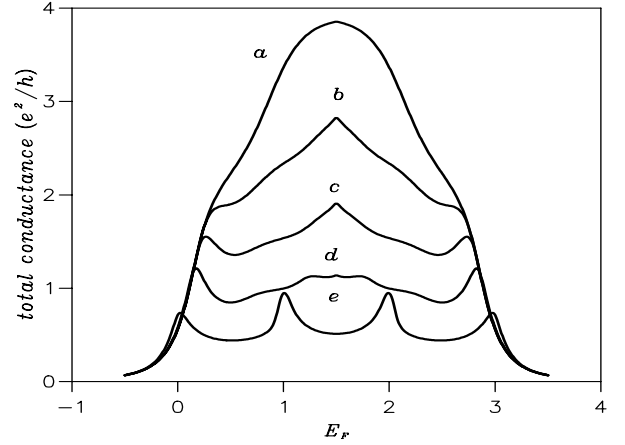


FIG. 2. Total conductance for the same case of Fig. 1 as a function of  $E_F$  for different temperature values  $T/U$ : a) 0.0005, b) 0.0025, c) 0.005, d) 0.01 and e) 0.03.

So far we have analyzed the case of a completely degenerate level. In an actual QD geometrical asymmetries could result in an effective splitting  $\Delta E$  of the dot levels. When  $\Delta E \gg \Gamma$  the physical situation could be described by a single level Anderson model. It is interesting to analyze the evolution from this situation to the quasi-degenerate case  $\Delta E \rightarrow 0$  previously discussed. This evolution is depicted in Fig. 3 and 4 where a splitting  $\Delta E = 0.1U$  and  $\Delta E = 0.5U$  is introduced. Only the first half of the charging curve is shown as the total conductance is symmetrical with respect to  $E_F = (3U + \Delta E)/2$ . As can be observed in the charging curves shown as insets in these figures, the splitting tends to block the charging of the upper levels. This blocking effect is nearly complete for  $\Delta E = 0.5U$  and is already apparent in the case of Fig. 3 where  $\Delta E \sim \Gamma$ . The total conductance at low temperatures (case a) is observed to decrease for increasing splitting approaching the behavior  $2(e^2/h) \sin^2[\pi n_m]$ , where  $m$  denotes the lower level. The overall effect of temperature is again to reduce the total conductance in the range for  $E_F$  where the Kondo effect is present. In the case of Fig. 4 the conductance *increases* with temperature in the region  $E_F \sim 1.5U$  where the higher level starts to be populated. One can also notice for this case that the conductance peaks associated with the lower level are not completely symmetrical due to the influence of the second level. Both effects, the increase of conductance with temperature between each pair of peaks as well as

the asymmetry, are present in the experimental results of Ref.<sup>1</sup> and can be considered as a manifestation of the multilevel structure of an actual QD.

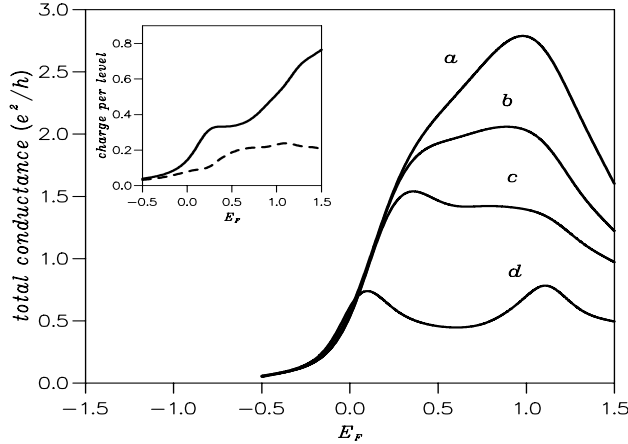


FIG. 3. Total conductance for a two-level dot with splitting  $\Delta E/U = 0.1$  as a function of  $E_F$  for different temperature values  $T/U$ : a) 0.0005, b) 0.0025, c) 0.005 and d) 0.03. The inset shows the charge per spin on the two levels as a function of  $E_F$ . Only the first half of the curve is shown as the total conductance is symmetrical with respect to  $(3U + \Delta)/2$ .

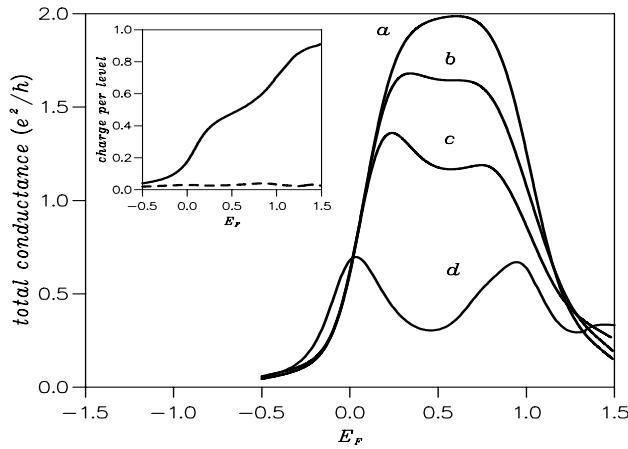


FIG. 4. Same as Fig. 3 for  $\Delta E/U = 0.5$ .

The most remarkable consequences of this multilevel structure would be observed in a situation corresponding to a smaller splitting where “collective” Kondo like features like the ones depicted in Figs. 2 and 3 should appear. Vertical dots with cylindrical symmetry, as those studied in Ref.<sup>19</sup>, constitute an almost ideal realization of the two-fold degenerate case. One word of caution should be said, however, regarding the effects of Hund’s rule, not included in the present approach and which might introduce some deviation with respect to the behavior depicted in Fig. 2. The results presented in this letter will fully apply to a situation where the exchange interaction between the degenerate levels is much smaller than

the Coulomb interaction  $U$ .

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